HIGH FIELD MAGNETORESISTANCE IN $HfTe_5$ AND $ZrTe_5$

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Abstract----Transverse magnetoresistance of $HfTe_5$ and $ZrTe_5$ was measured. Shubnikov-de Haas oscillation at low magnetic field enabled us to determine a cyclotron mass $m_{cyc.}/m_0$ (m_0; free-electron mass) = 0.02 (0.04) and a Dingle temperature T_D = 4.5 K (2.7 K) in $HfTe_5$ ($ZrTe_5$). The magnetoresistance at the field of the quantum limit showed a large peak accompanied by an extremely deep resistance minimum at the higher field side. This anomaly was explained qualitatively by a magnetically induced band-crossing model.

INTRODUCTION

Since the observation of the strong resistivity peak in the transition-metal pentatellurides: $HfTe_5$ and $ZrTe_5$, intensive studies have been done for searching the mechanism of the resistivity anomaly [1-3]. The crystal structures of $HfTe_5$ and $ZrTe_5$ are almost identical and have been classified as the $HfTe_5$ type structure by Hulliger [4]. Each transition-metal atom in $HfTe_5$ has essentially the same coordination as in the type A-$ZrSe_3$ structure [5]. Since the metal-chalcogen linkage between $ZrSe_3$-type chains along the a axis creates a set of layers, the physical properties of these materials are expected to have one- or two-dimensional characteristics. Existence of unusual resistivity peak in these pentatellurides led us to determine the form of the Fermi surface below the temperature of the resistivity anomaly, T_p. In the previous reports we have found out the Shubnikov-de Haas (SdH) oscillations below 4.2 K [6,7]. In both compounds single cross-sectional frequency dominates the oscillatory behavior, and their angular dependence clearly shows the existence of a long cigarette-shaped ellipsoidal Fermi surface, reflecting quasi-two-dimensional character in the a-c crystallographic plane of the crystal [8]. In this paper, we report the low-field (up to 2.5 T) and high-field (up to 36 T) transverse magnetoresistance. Effective cyclotron mass and Dingle temperature are determined for $HfTe_5$ and $ZrTe_5$ in the configuration in which the direction of the magnetic field is parallel to the b axis. Anomalous magnetoresistance peak was observed around 2.5 T in $HfTe_5$ and 8 T in $ZrTe_5$ and it is qualitatively explained by a magnetically induced band-crossing model.

EXPERIMENTAL DETAIL

The temperature dependence of the SdH oscillations in $HfTe_5$ and $ZrTe_5$ was measured in the range from 0.32 to 4.2 K using a ^3He cryostat system which includes an 8 T superconducting solenoid. The dc transverse magnetoresistance measurement was performed by a standard four-probe method. The direction of the field was parallel to the b axis. The four electrodes were attached by a silver paste (DuPont 4929). Constant current (0.2 - 0.5 mA) was supplied parallel to the chain a axis of the crystal.

Discharge from a 5 mF, 5000 V capacitor bank produces high pulsed magnetic field in a coil of normal conductor cooled in liquid nitrogen. The field, which is approximately a half sine wave, has a peak value of 36 T, and lasts 5 msec. Transverse magnetoresistance was measured in the same configuration as that in the low field measurement mentioned above. The sample was fixed on a circuit board. The lead wires were formed by etch stripes to avoid mechanical vibration of the sample and leads under the pulse field as is illustrated in Fig. 1. The contacts were made by indium solder.

RESULTS AND DISCUSSION

Figure 2 shows the low-field SdH oscillatory component of the dc magnetoresistance (plotted by the open circles) in $HfTe_5$ (a) and $ZrTe_5$ (b). As is seen in the figure, single cross-sectional frequency of 0.12 T (0.54 T) dominates the oscillation in $HfTe_5$ ($ZrTe_5$). No spin splitting was observed. The values of the cyclotron mass $m_{cyc.}$ in the a-c plane and Dingle temperature T_D were estimated by fitting the oscillatory component of the transverse magnetoresistance to the following expression [9],

$$\frac{\Delta \rho}{\rho_0} = A \gamma \cos\left(\frac{2\pi E_F}{\hbar \omega_{cyc.}} - \phi\right), \tag{1}$$

where A is a constant scale factor, $\omega_{cyc.}$ is a cyclotron frequency in the a-c plane and E_F is Fermi energy. γ is given by

$$\gamma = -\left(\frac{\hbar \omega_{cyc.}}{2 E_F}\right)^{1/2} \frac{2\pi^2 kT/\hbar\omega_{cyc.}}{\sinh(2\pi^2 kT/\hbar\omega_{cyc.})} \exp\left(-\frac{2\pi^2 k T_D}{\hbar \omega_{cyc.}}\right). \tag{2}$$

Here, we neglected the effect of spin and of higher harmonics. We also neglected the temperature dependence of E_F and of T_D. The best fit to the data at 0.32, 1.2, and 4.2 K is $m_{cyc.}/m_0$ = 0.02 (0.04) and T_D = 4.5 K (2.7 K) for $HfTe_5$ ($ZrTe_5$), where m_0 is the free-electron mass. Fitted curves are shown by solid lines in Fig. 2.

It is worth noticing that the spacing between the leftmost peak and the vertical axis in Fig. 2b is apparently smaller than the period of the oscillation, which cannot occur in a simple one-band scheme in which n=0 Landau level is unattainable for the Fermi level. This unusual phase shift will be explained by a magnetically induced band-crossing model discussed later.

As shown in Figs. 3 and 4, the magnetoresistance curves for $ZrTe_5$ and $HfTe_5$ at 4.2 K are similar in some respects.

SdH oscillation is maintained up to about 8 T for $ZrTe_5$; the corresponding field for $HfTe_5$ is about 2.5 T. Around these fields, both curves have a peak and a dip which are so large and insensitive to temperature that they should be distinguished from the oscillation. The dip appears to be especially deep in $ZrTe_5$. The resistance at the bottom of the dip is sample dependent ranging from -10 to +30 % of the zero-field value, the negative magnetoresistance was observed only in one of the five samples measured, however. As shown in Fig. 4, the peak and the dip change their magnitude very moderately with temperature. Shifting their positions to the higher fields, they continue to exist from 4.2 to 100 K. Therefore, we may safely conclude that they come from an anomaly which involves the change in the background behavior rather than that in the SdH oscillation.

It is hard to attribute this anomaly to the behavior of a single band in a magnetic field. The Hall experiment [3,10] also suggests that at least two electron bands contribute to the conduction below T_p. So we examined a simple model that involves the competition for carrier population between two electron bands: one with light cyclotron mass (band 1 in Fig. 5) and the other with heavy one (band 2 in Fig. 5). In the presence of a magnetic field, the states in band 1 are quantized, and the density of states $D_1(E)$ is given by [11]

$$D_1(E) = \frac{eBm_z^{1/2}}{2\pi^2\hbar^2 c} \times \sum_{n=0}^{\infty} \left[\frac{E-(n+\frac{1}{2})\hbar\omega_{cyc.} + \{(E-(n+\frac{1}{2})\hbar\omega_{cyc.})^2+\Gamma^2\}^{1/2}}{\{E-(n+\frac{1}{2})\hbar\omega_{cyc.}\}^2+\Gamma^2} \right]^{1/2}, \qquad (3)$$

where E is the energy, n is the quantum number of Landau level, and m_z is the effective mass of band 1 along the magnetic field B. We included the effect of collision broadening through the factor Γ ($=\pi kT_D$). For band 2, we assumed that the effective mass is isotropic and so heavy that we can write the density of states $D_2(E)$ in the classical form

$$D_2(E) = \frac{1}{2\pi^2}\left(\frac{2m_2}{\hbar^2}\right)^{3/2}\sqrt{E - E_G}\;, \qquad (4)$$

where m_2 is the effective mass of band 2 and E_G is the energy difference of the edges of the two bands at B=0.

As an applied magnetic field is intensified, the redistribution of carriers, which arise from the diamagnetic energy shifts of the bands, takes place so that the Fermi levels of the two bands are equated. The total electron density N is fixed as is expected from the result of the Hall experiment [10]. Since the cyclotron mass of band 1 is lighter than that of band 2, the energy of band 1 increases faster than that of band 2 (Fig.5). Finally at a certain field, when the edge of the band 1 crosses the Fermi level of the system, band 1 becomes almost vacant.

We illustrated the calculated change of the electron density N_1 in band 1 in Fig. 6. The parameters of band 1 were taken from the result of the SdH experiment and the unknown effective mass of band 2 was assumed to be m_0. The parameters are given in Table I. This figure explains qualitatively the magnetoresistance data in the following way.

1) At relatively low field (< 8 T), both band 1 and band 2 are populated. The cyclotron mass of the carriers in band 2 are so heavy that band 1 mainly contributes to the magnetoresistance. Band 1 shows the SdH oscillation. The resistance may be large in its magnitude due to the two-carrier effect.
2) At high field (> 15 T), only band 2 is populated. Moderate magnetoresistance associated with the heavy carrier is observed.
3) At intermediate field (8 < B <15 T), the system is at the transition from two-carrier region to one-carrier region. The magnetoresistance decreases remarkably.

In Fig. 6, the population of band 1 changes rather drastically with magnetic field. But this does not disturb the SdH period. The graph of Fig. 7 represents the Fermi energy and some of Landau levels in band 1 at different magnetic fields. Intersections of these lines are equally spaced in 1/B plot as in Fig. 8. Since the Fermi energy changes almost linearly with B, the period of oscillation remains unaffected. It is just the phase that is affected. The Fermi level crosses n=Ø Landau level at finite intensity of magnetic field (Fig. 8). This means that an extra phase is induced from the argument $2\pi E_F/\hbar\omega_{cyc.}$ in Eq. (1) besides the phase ϕ in one-carrier case.

In conclusion, the transverse magnetoresistance of $HfTe_5$ and $ZrTe_5$ is characterized by the SdH oscillation with single cross-sectional frequency, and by the anomalous peak and dip. Analysis on SdH oscillation at low temperature showed that the effective cyclotron mass is Ø.Ø2$m_Ø$ in $HfTe_5$ (Ø.Ø4$m_Ø$ in $ZrTe_5$) and the Dingle temperature is 4.5 K in $HfTe_5$ (2.7 K in $ZrTe_5$). The anomalous peak and dip were partially explained by a magnetically induced band-crossing model.

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TABLE I. Parameters used in the calculations

total electron density	6.5×10^{17} cm^{-3}
energy difference of the two bands (E_G)	13.4 meV
cyclotron mass in band 1 ($m_{cyc.}$)	$0.04m_0$
effective mass in band 1 (m_z)	$2.7m_0$
effective mass in band 2 (m_2)	m_0
broadening factor ($\Gamma = \pi kT_D$)	0.731 meV
temperature	0 K

FIGURE CAPTIONS

Fig. 1 Schematic picture of sample holder used in the pulsed magnetic field measurement.

Fig. 2 Oscillatory component of dc magnetoresistance in $HfTe_5$(a) and $ZrTe_5$(b). The circles are the experimental results and the solid curves are the best fitting by the calculation according to Eq. (1).

Fig.3 Dc transverse magnetoresistance in $HfTe_5$ at 4.2 K.

Fig.4 Dc transverse magnetoresistance and its temperature dependence in $ZrTe_5$.

Fig.5 Magnetically induced band-crossing model.

Fig.6 Calculated change of ratio of the electron density N_1 in band 1 to the total electron density N using the parameters given in Table I.

Fig.7 Calculated changes of the Landau levels and the Fermi level in band 1.

Fig.8 Calculated changes of the Landau levels and the Fermi level in band 1 plotted against reciprocal magnetic field.

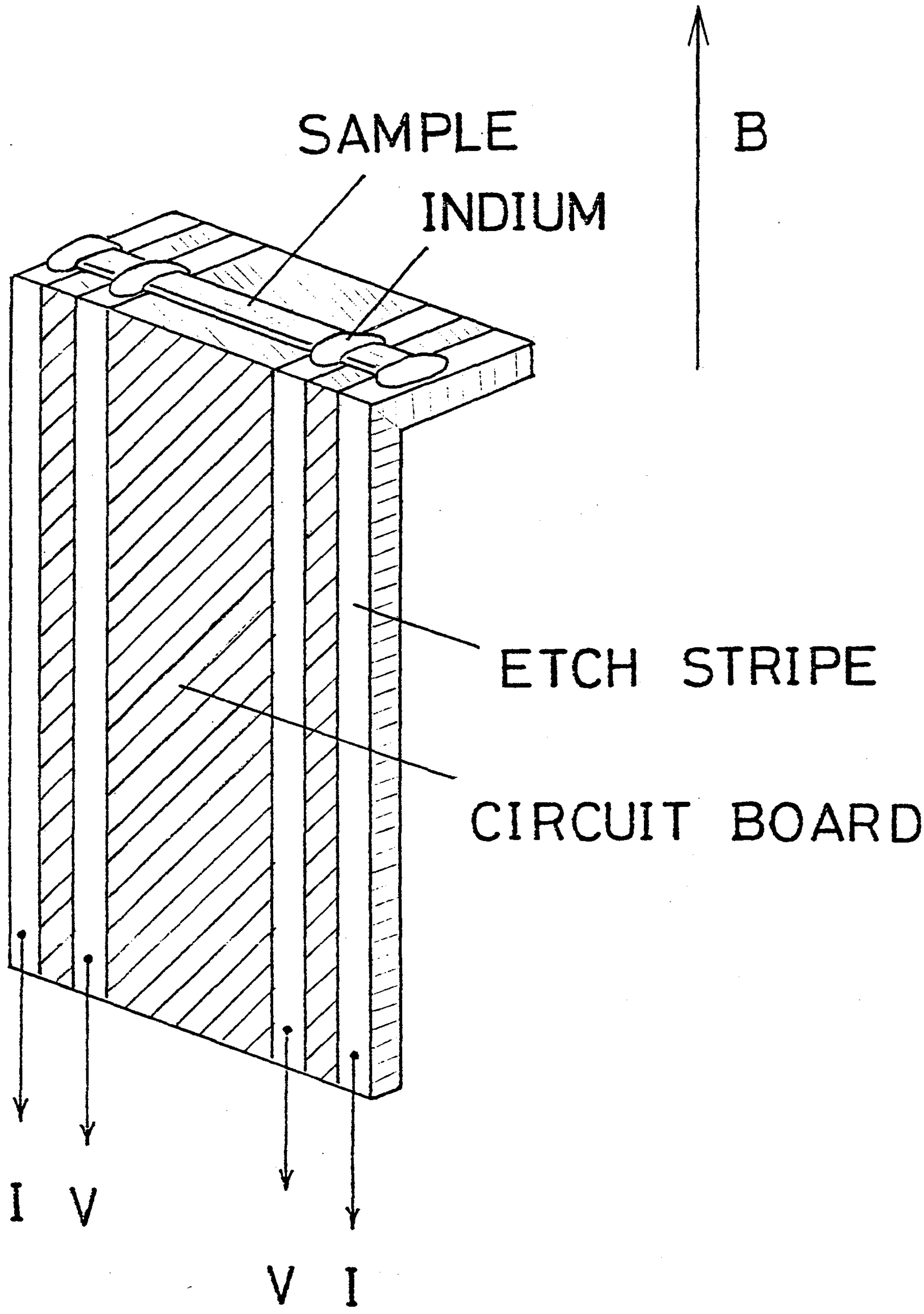


Fig. 1

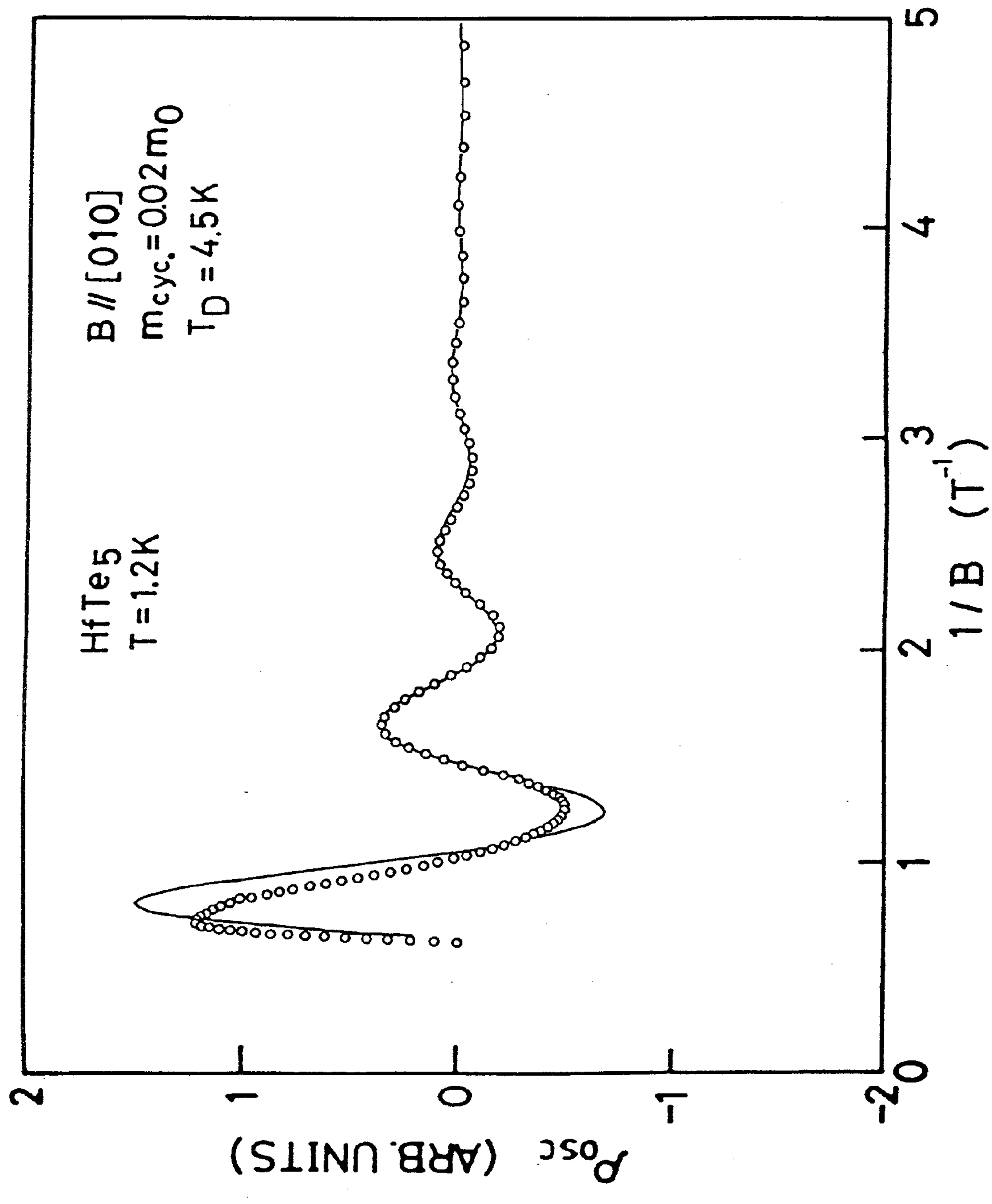


Fig. 2 a

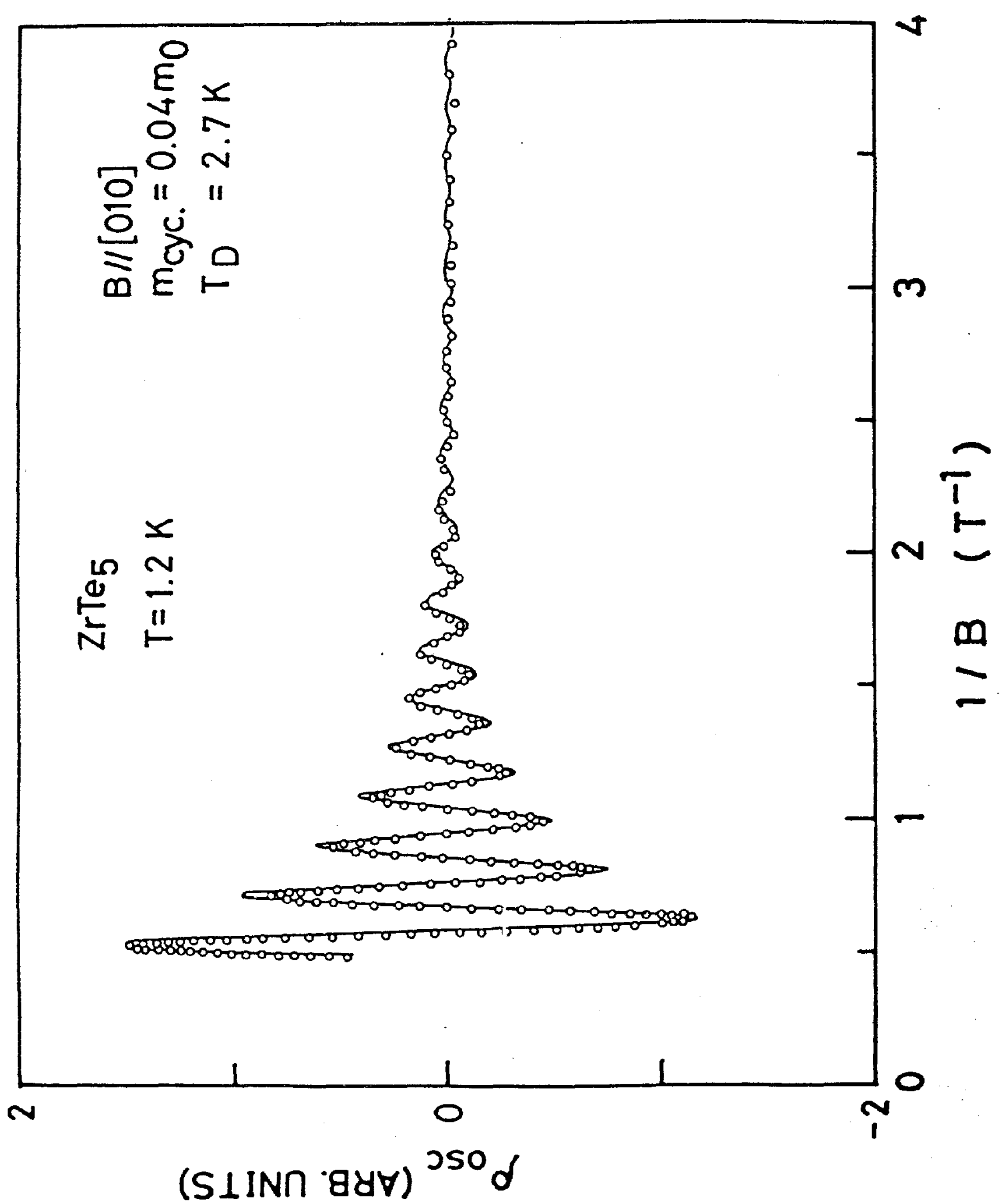

ZrTe5
T=1.2 K
B//[010]
mcyc. = 0.04m0
TD = 2.7 K
1/B (T-1)
ρosc (ARB. UNITS)
2
0
-2
0
1
2
3
4

Fig. 2 b

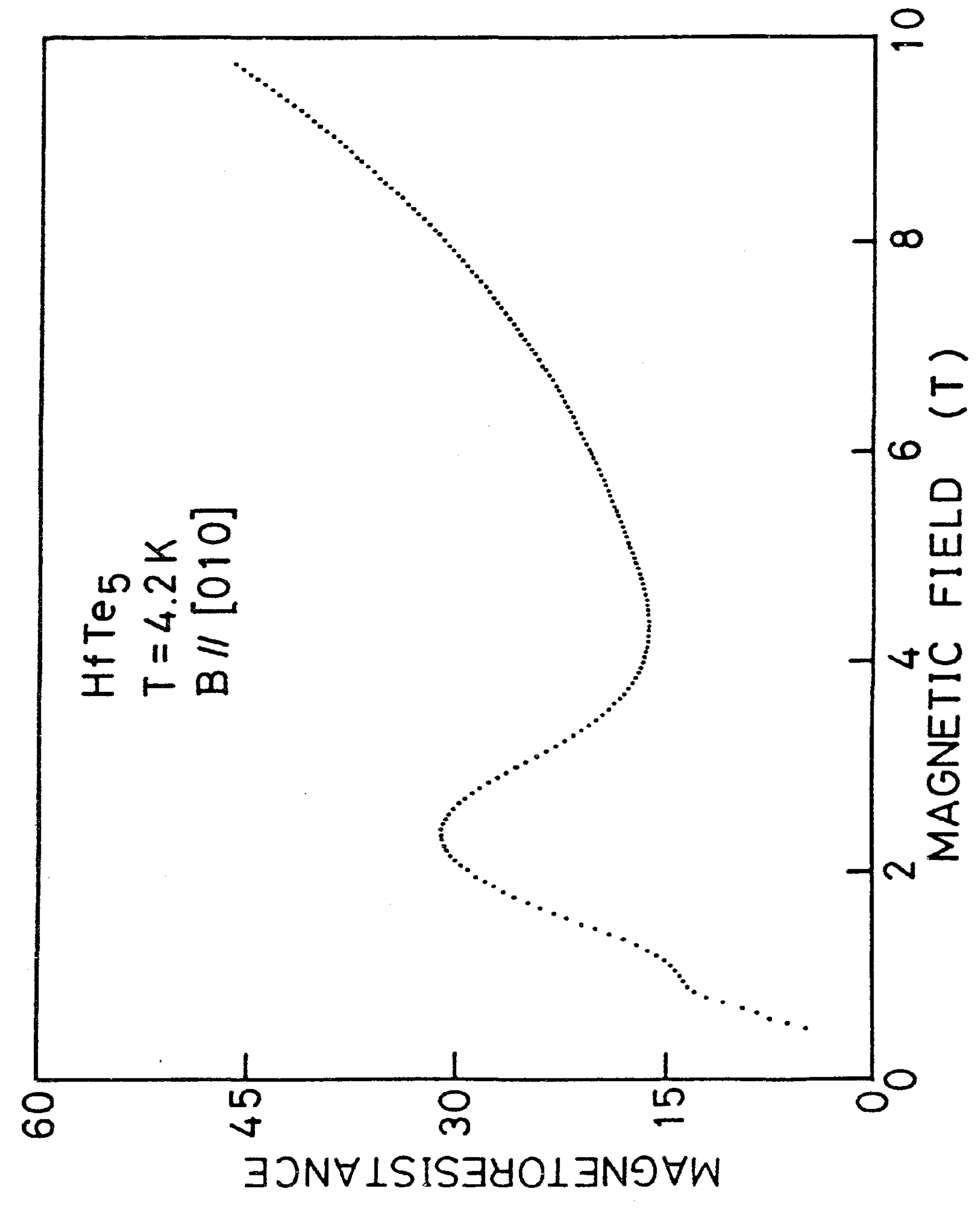
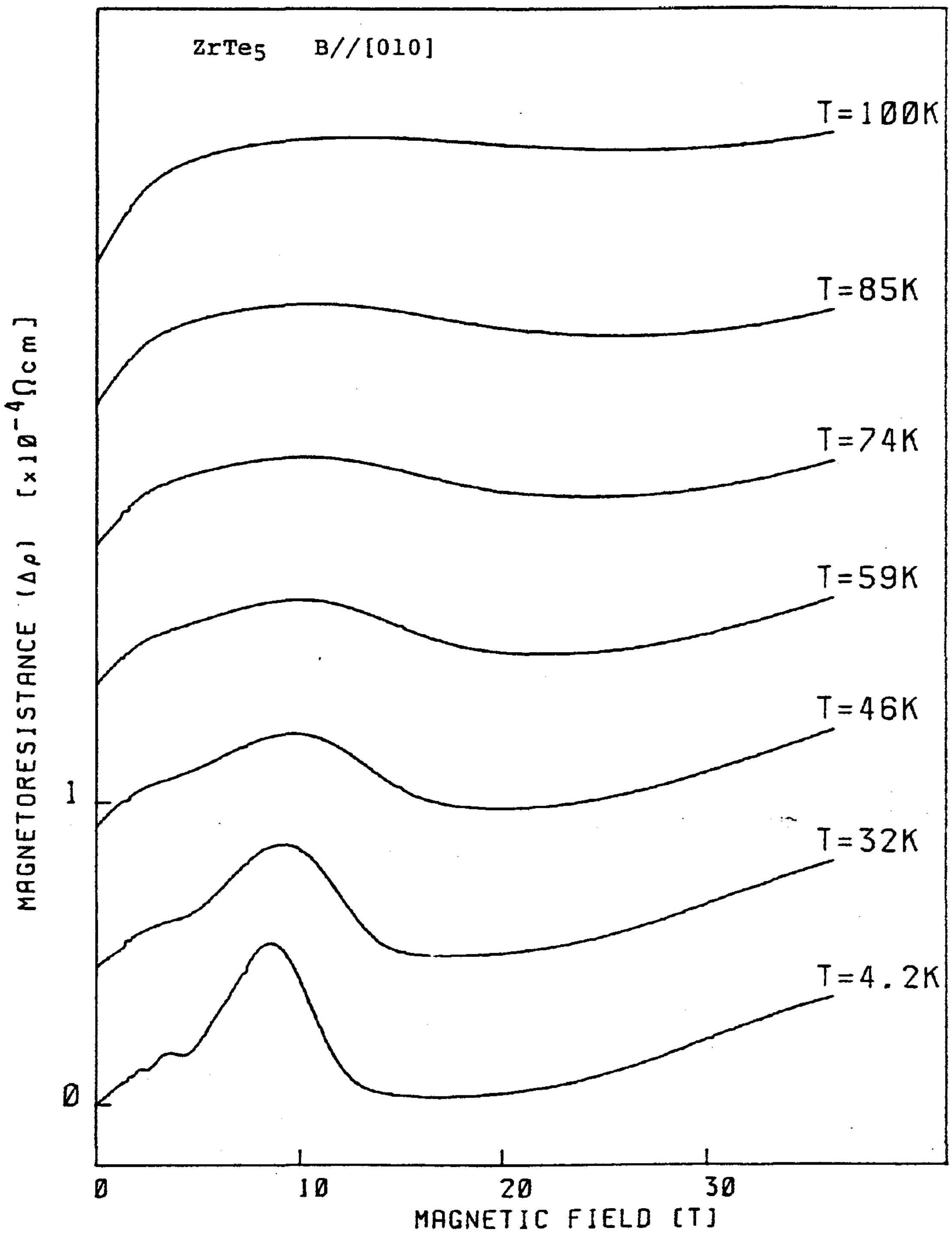

HfTe$_5$
T = 4.2 K
B // [010]
MAGNETIC FIELD (T)
MAGNETORESISTANCE
0
2
4
6
8
10
15
30
45
60

Fig. 3



ZrTe5 B//[010]
T=100K
T=85K
T=74K
T=59K
T=46K
T=32K
T=4.2K
MAGNETORESISTANCE (Δρ) [x10^-4 Ωcm]
1
0
0
10
20
30
MAGNETIC FIELD [T]

Fig. 4

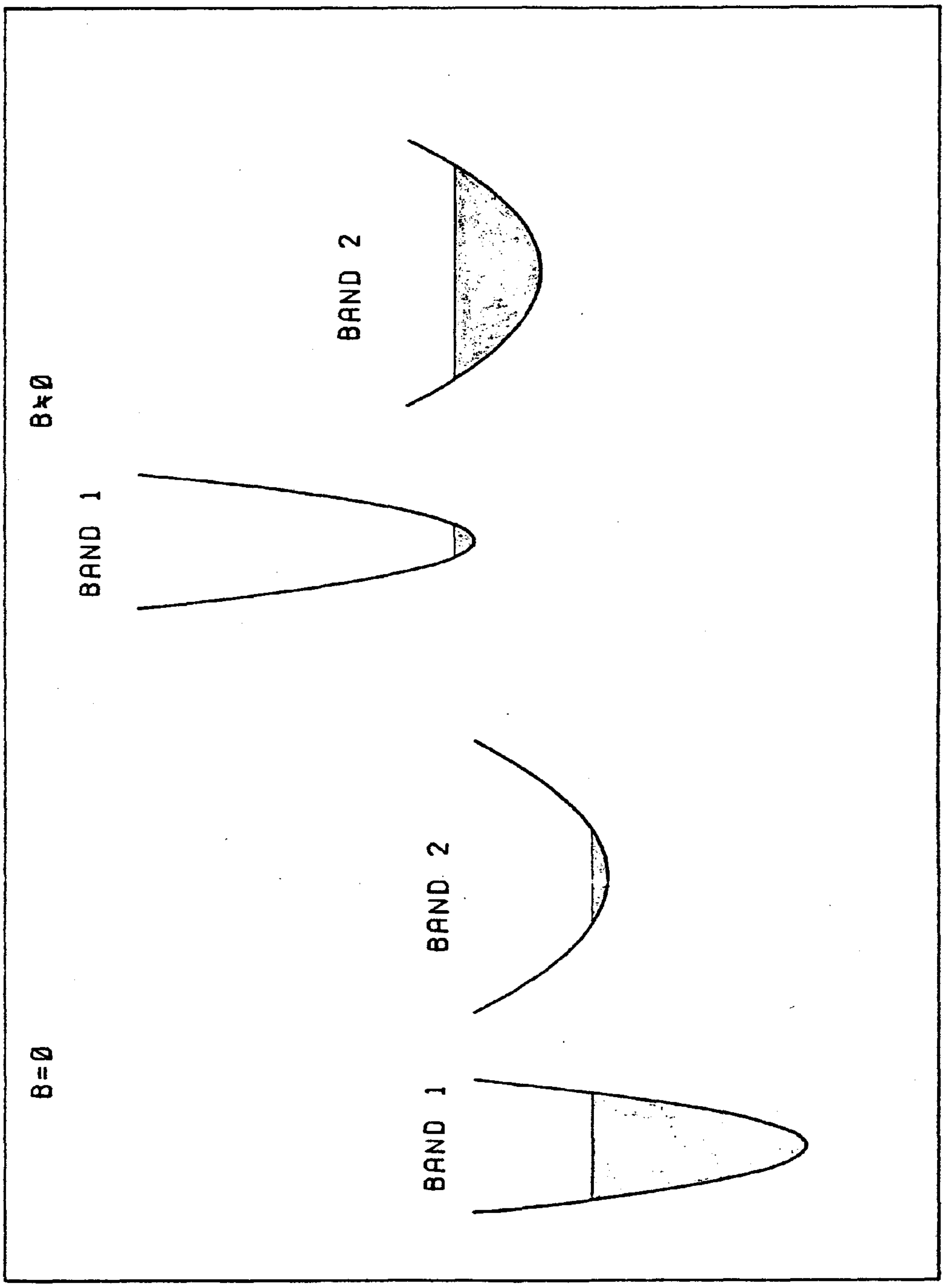


Fig. 5

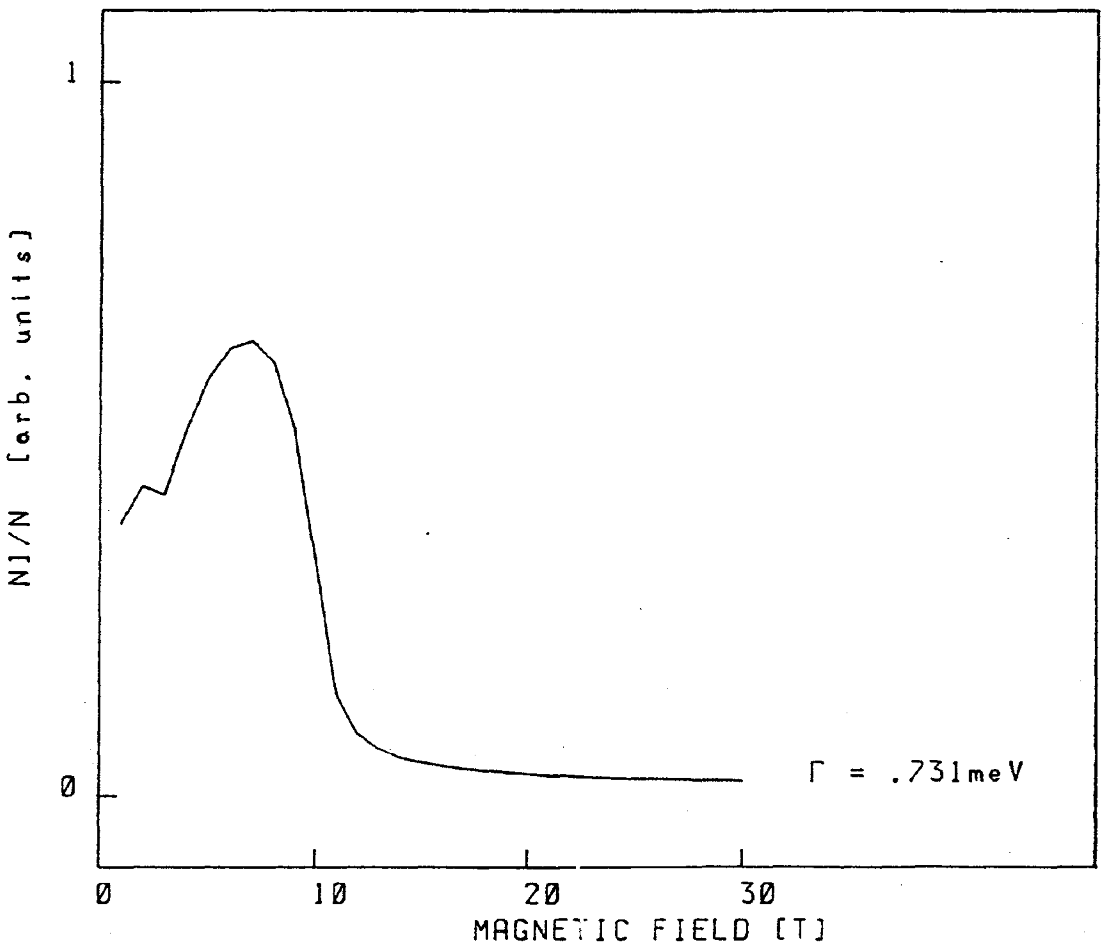
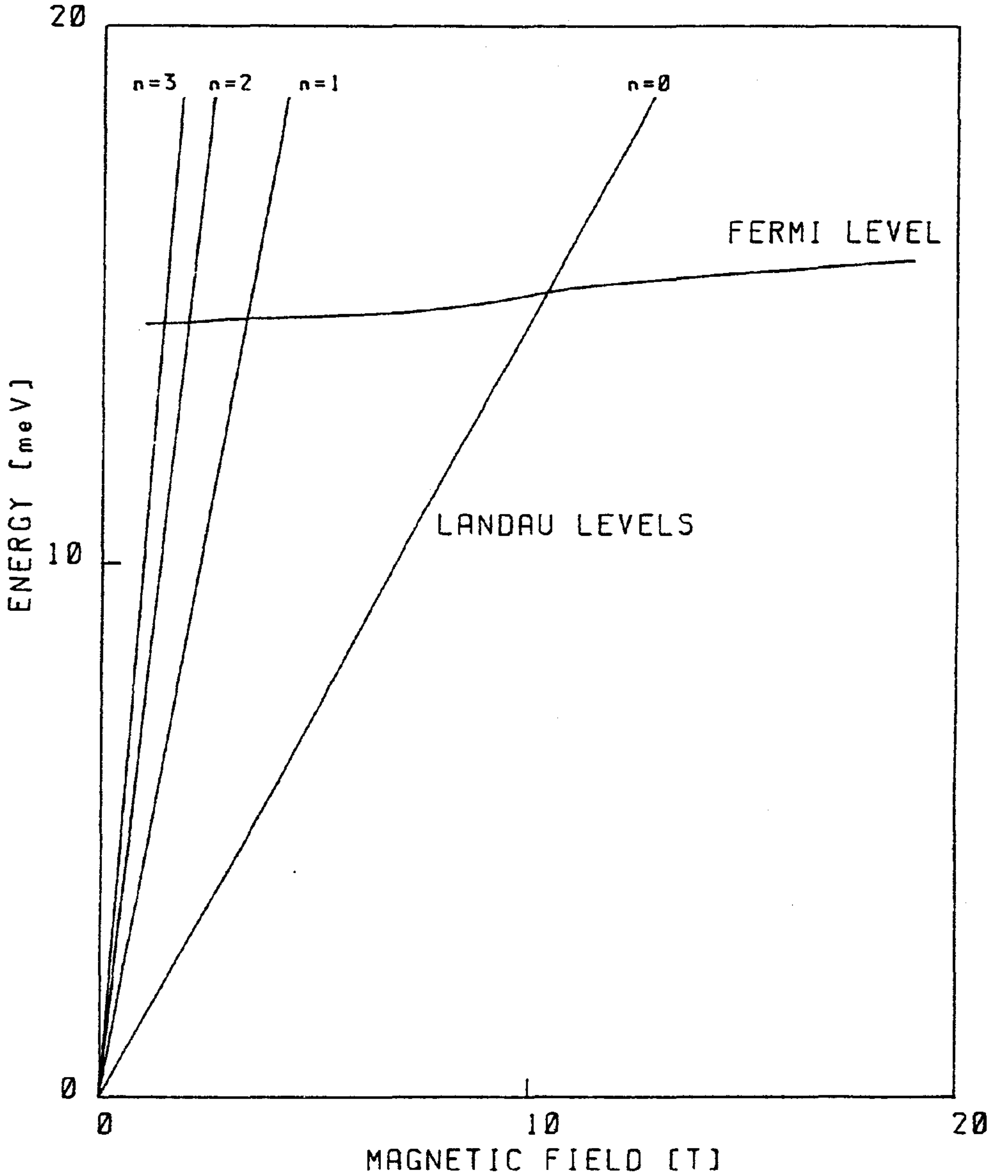


Fig. 6



20
n=3
n=2
n=1
n=0
FERMI LEVEL
ENERGY [meV]
10
LANDAU LEVELS
0
0
10
20
MAGNETIC FIELD [T]

Fig. 7

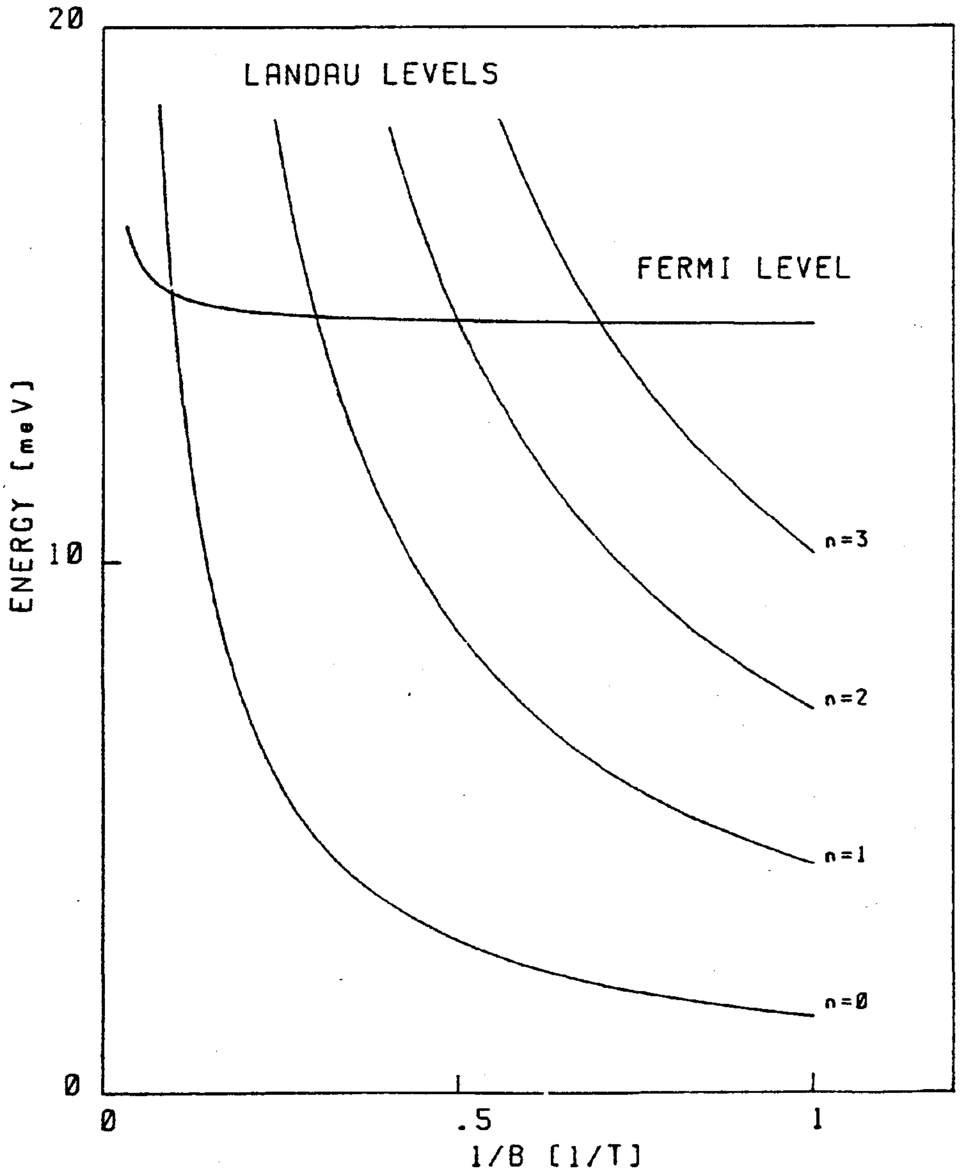

LANDAU LEVELS
FERMI LEVEL
n=3
n=2
n=1
n=0
ENERGY [meV]
20
10
0
0
.5
1
1/B [1/T]

Fig. 8